Nonexponential Relaxation of Magnetization at the Resonant Tunneling Point under a Fluctuating Random Noise

Seiji MIYASHITA and Keiji SAITO
Department of Applied Physics, Graduate School of Engineering,
University of Tokyo, Bunkyo-ku, Tokyo 113-8656, Japan
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Nonexponential relaxation of magnetization at resonant tunneling points of nanoscale molecular magnets is interpreted to be an effect of fluctuating random field around the applied field. We demonstrate such relaxation in Langevin equation analysis and clarify how the initial relaxation (square-root time) changes to the exponential decay. The scaling properties of the relaxation are also discussed.

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As to the relaxation of metastable magnetization of uniaxial nanoscale molecular magnets such as Mn_{12} and Fe_8, the resonant tunneling phenomena have been paid attention and various interesting properties of the phenomena have been reported [1–9]. Properties of the phenomena have been also investigated in the theoretical side [10–20]. We have studied such relaxation from a view point of nonadiabatic transition among their discrete energy structure due to a finite number of degrees of freedom. There we studied the relation between the relaxation rate and the sweeping velocity. Actually such relation has been utilized to estimate the energy gap in a recent experiment [8,9]. Because the tunneling gap is so small, thermal environments have strong influence on the tunneling, which has been studied extensively.

In the case of static field, there have been many experiments reporting that nonexponential relaxation occurs at the resonant point. That is, the initial relaxation can not be fitted by usual exponential function but is fitted well by a stretched exponential function with the exponent near 0.5 or a square-root function [7,21–23]. If the field would be precisely fixed at the resonant point, we should see the coherent tunneling. Thus the observations indicate that the decoherent effect plays an important role. This nonexponential relaxation has been interpreted as a phenomenon due to the fact that the region of the field of the resonant tunneling is very narrow and small fluctuation can detune the resonance condition by Prokof'ev and Stamp [14]. They considered the distribution of the internal field which mainly consists of dipolar field from other molecules, and investigated its time evolution. Combining the evolution of the distribution and the relation between the steady state distribution of the dipolar field and the magnetization,they found a square-root time initial relaxation and an exponential relaxation in the late stage, which explains the overall dependence of the experiments.

In the detailed observation on the distribution of internal field $P(\xi)$ in Fe_8 [9], the square-root dependence is found even if the initial magnetization is zero, and change of the distribution appears only locally and it is not associated with the reforming total distribution of $P(\xi)$. These features are not compatible with the mean-field type analysis of $P(\xi)$, and thus it seems necessary to consider more general mechanism of the square-root time initial relaxation. Actually it has been pointed out [24] that the fast fluctuation of hyperfine fluctuations are very important in the local field dynamics. In this Letter, we propose an alternate explanation of this phenomenon as a general phenomenon at narrow resonant points with fluctuating field using a Langevin equation approach, i.e., using the Ornstein-Uhlenbeck process [25].

Here we consider a two-level system for the simplicity and its Hamiltonian is given by

$$\mathcal{H} = h_{\rm ext}(t)\sigma^z - \frac{\Delta E}{2}\sigma_x, \tag{1}$$

where σ_x and σ_z are the Pauli matrices and $h_{\rm ext}(t)$ is a time-dependent external field, and $\frac{\Delta E}{2}$ denotes the transverse field which represents the quantum fluctuation of the z component of the spin. We solve the Schrödinger equation of this system with time dependent field $h_{\rm ext}(t)$ by applying the time evolution function

$$|t+\Delta t\rangle = \exp(-i\mathcal{H}(t))|t\rangle, \tag{2}$$

where the $h_{\rm ext}(t)$ is prepared by a Langevin equation.

Corresponding to the molecules of Mn_{12} or Fe_8, we consider that the resonant point is very narrow and is regarded as a point. The external field consists of the static part h_0 and a fluctuating part $h(t)$:

$$h_{\rm ext}(t) = h_0 + h(t). \tag{3}$$

The fluctuating part is caused by independent changes of many magnetizations around the site. Thus we consider that this part is an assembly of independent fluctuation

$$h(t) = h(0) + \int_0^t \sum_j \delta h_j(s)ds, \tag{4}$$

where $\delta h_j(s)$ is the change of a field from the j-th site (h_j) at time s. Here we assume that $\delta h_j(s)$ is independent of time and position, and we regard it as a white gaussian noise $\eta(s)$

$$\langle \eta(t) \rangle = 0, \quad \langle \eta(t)\eta(s) \rangle = 2D\delta(t-s). \tag{5}$$

Here the assumption of the gauss distribution is not essential but we assume it for the convenience of analysis. Then, the relation(4) is written as

$$h(t) = h(0) + \int_0^t \eta(s) ds = W(t), \tag{6}$$

where W is the Wiener process

$$\langle W(t) \rangle = 0, \quad \langle W(t)W(s) \rangle = 2D\mathrm{Min}(t,s). \tag{7}$$

The distribution of $W(t)$ is given by

$$P(W) = \frac{1}{\sqrt{4\pi Dt}} \exp\left(-\frac{W^2}{4Dt}\right). \tag{8}$$

We show an example of the Wiener process in Fig. 1 (a dashed line).

The nonadiabatic transition only occurs when the field crosses the resonant point. Hereafter we set the resonant point at $h = 0$ and also the static field at $h_0 = 0$. The probability p of the adiabatic transition of the state, i.e., from the ground state to the ground state (or from the excited state to the excited state), is given by the Landau-Zener-Stückelberg (LZS) formula [26–28]

$$p = 1 - \exp\left(-\frac{(\Delta E)^2}{2|M - M'|v}\right), \tag{9}$$

where ΔE is the energy gap at the crossing point (tunnel gap), M and M' are the magnetizations of crossing states, and v is the speed of the field at the resonant point. The transition probability from the initial distribution (p_1, p_2) to the scattered distribution (p_1', p_2') depends on the velocity v of the field at the time of crossing and also on the phase factor due to the free motions outside of the crossing region [29]. Here we consider many sample of $\{h_{\rm ext}(t)\}$ and the ensemble average over the distribution of v and the phase factor.

The change of magnetization ($M \to M'$) at this crossing is given by $M = M_0(p_1 - p_2) \to M' = M_0'(p_1' - p_2')$, where M_0 and M_0' are the magnetization of the each ground state. In Fig. 1, we show a dynamics of magnetization $M(t)$ for the shown process of $h_{\rm ext}$.

From the dependence of (9), the average probability of the adiabatic transition, which changes the magnetization at a crossing, is estimated as

$$p_{\rm av} \simeq \langle \frac{(\Delta E)^2}{2|M - M'|v} \rangle = \alpha_0 \frac{(\Delta E)^2}{\sqrt{D}}, \tag{10}$$

where we assumed that the value of p is small and that the average velocity of the field is proportional to the strength of the random field $\sqrt{D}$. The change of magnetization at a crossing is given by $M' = (1 - 2p_{\rm av})M$.

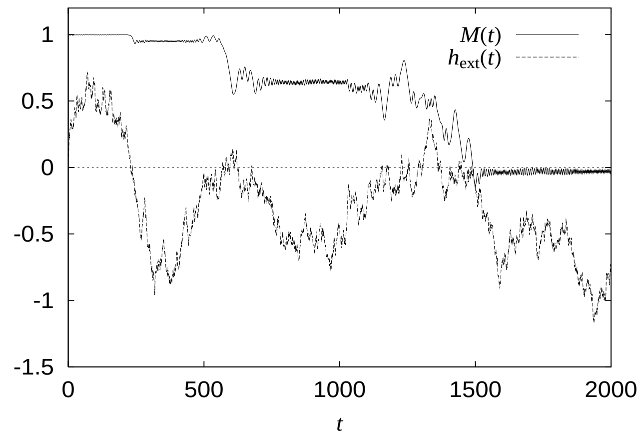

FIG. 1. A sample of the Wiener process (dashed line) and the change of the magnetization for this process of $h_{\rm ext}$.

If we assume that the sequence of the crossings give independent contributions, the change of magnetization for arbitrary initial value $M(0)$ is given by

$$M(t) = M(0)(1 - 2p_{\rm av})^{N_{\rm ac}(t)}, \tag{11}$$

where $N(t)_{\rm ac}$ is the accumulated number of the crossings by the time of t.

The number of crossings per unit time, $N(t)$, is estimated from the probability for $|W| \le \sqrt{D}$, where $\sqrt{D}$ is the jump range of the field. Thus $N(t)$ at time t is given by

$$N(t) \propto \frac{1}{\sqrt{4Dt}} \times \sqrt{D} \propto \frac{1}{\sqrt{t}}. \tag{12}$$

Thus the accumulated number of crossings by the time t is given by

$$N_{\rm ac}(t) = \int_0^t N(t) dt = c\sqrt{t}, \tag{13}$$

where c is a constant. Thus the total change of the magnetization is

$$\begin{aligned} M &= M_0(1 - 2\alpha_0 \frac{(\Delta E)^2}{\sqrt{D}})^{c\sqrt{t}} \\ &\simeq \exp\left(-\alpha \frac{(\Delta E)^2}{\sqrt{D}} \sqrt{t}\right), \end{aligned} \tag{14}$$

where $\alpha = 2\alpha_0 c$. This constant is uniquely determined by the nature of random process which will be determined later. Thus we conclude that the magnetization shows a stretched exponential decay with the exponent 1/2, which shows the square-root time initial relaxation. In Fig. 2, we show the averaged magnetization $\langle M(t) \rangle$ over the 10,000 samples of $h_{\rm ext}$. In the inset $M(t)$ is plotted in the coordinate $(\sqrt{t}, \log M(t))$, where we confirm the dependence of (14). This dependence gives the initial square-root time dependence

$$M = M_0(1 - \alpha \frac{(\Delta E)^2}{\sqrt{D}} \sqrt{t}). \quad (15)$$

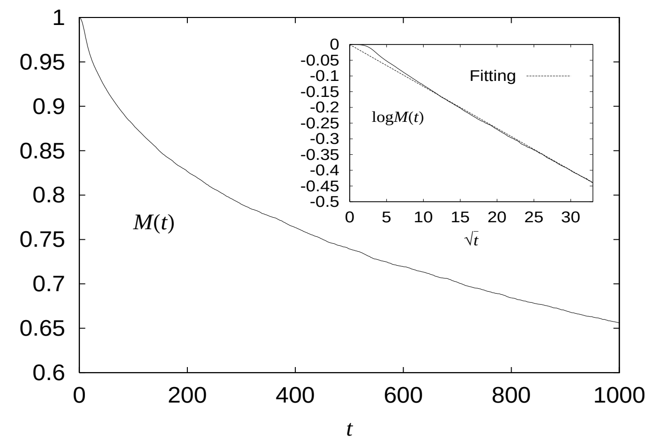

FIG. 2. The averaged magnetization $\langle M(t) \rangle$ over the 10,000 samples of $h_{\rm ext}$. The inset shows a plot $(\log M(t), t^{1/2})$, where the dotted line is a straight guide line for eye.

In the real situation the field $h(t)$ may not deviate without limit and there are some restoring mechanisms. Next we study the mechanism of transition from the initial relaxation (14) to the exponential relaxation. Taking into account the restoring effect, we adopt the Ornstein-Uhlenbeck process for the evolution of $h(t)$:

$$\frac{dh}{dt} = -\gamma h(t) + \eta(t). \quad (16)$$

The process (6) corresponds to the case $\gamma = 0$. The distribution of $h(t)$ for this process is given by

$$P(h) = \frac{1}{\sqrt{2\pi\sigma^2}} \exp\left(-\frac{(h - \langle h \rangle)^2}{2\sigma^2}\right), \quad (17)$$

where

$$\sigma^2 = \frac{D}{\gamma}\left[1 - \exp(-2\gamma t)\right]. \quad (18)$$

For $t \ll \gamma^{-1}$, this reproduces (8). On the other hand it has a stationary distribution $\sigma^2 = D/\gamma$ at large time $t \gg \gamma^{-1}$. At this late stage there is a constant probability that the $h(t)$ stays around 0, which causes a constant rate relaxation, i.e., the exponential relaxation. Therefore we expect

$$M = M_1 \exp\left(-\beta(\Delta E)^2 \sqrt{\frac{\gamma}{D}} t\right), \quad (19)$$

for a long time $t \gg (2\gamma)^{-1}$, while we have the relation (14) for a short time $t \ll (2\gamma)^{-1}$. Here β is a constant independent of Γ, γ, and D. M_0 and M_1 are constants corresponding to a kind of initial magnetization of each process, which does not coincide with the initial magnetization.

In Fig. 3(a) we show a time dependence of the magnetization $\langle M(t) \rangle$ averaged over 10,000 samples. In Figs. 3(b) and (c), we plot $M(t)$ in $(\sqrt{t}, \log M(t))$ and in $(t, \log M(t))$, respectively. There we find the crossover from the square-root time relaxation to the exponential relaxation around $t \sim (2\gamma)^{-1}$.

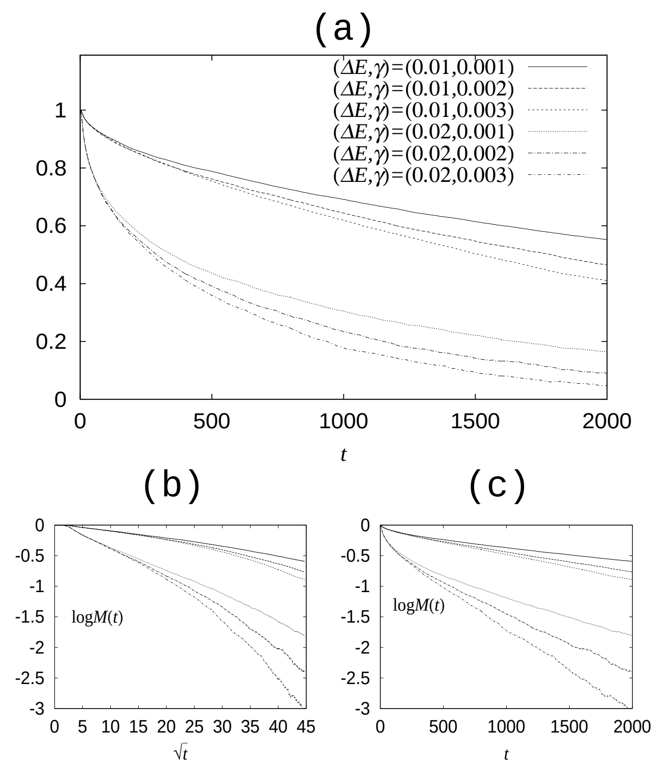

FIG. 3. Parameter dependence of $\langle M(t) \rangle$ for $h_{\rm ext}$ given by (16) for $D = 0.001$: (a) the linear plot $(t, \langle M(t) \rangle)$, (b) plot $(\sqrt{t}, \log\langle M(t) \rangle)$ for the initial relaxation (14), and (c) plot $(t, \log\langle M(t) \rangle)$ for the late relaxation (19).

In a very early time $t < 5$ we find a dead time where the magnetization does not change. This is considered to be due to the fact that the noise starting at 0 stays near the resonant region where the system essentially evolves coherently, i.e. $M(t) \simeq \cos(\Delta E t)$. This dead time becomes relatively short when ΔE and γ becomes small.

Studying $\langle M(t) \rangle$ for various parameters $(\Delta E, \gamma, D)$ we confirm that scaling relations of $M(t)$ on the parameters indicated by (14) and (19). In Figs. 4(a) and (b), we plot the data ($\Delta E = 0.01$, $\gamma = 0.001, 0.002$, and 0.003, and $D = 0.001, 0.002$ and 0.003) in the coordinates: $(\sqrt{t}, \log M(t)/((\Delta E)^2/\sqrt{D}))$ and $(((\Delta E)^2\sqrt{\gamma/D})t, \log M(t))$, respectively. We could plot the data in $(((\Delta E)^2/\sqrt{D})\sqrt{t}, \log M(t))$ instead of Fig. 4(a). However the scaling time region [the dead time $\sim O(1) < t < (2\gamma)^{-1}$] of each parameter set shown in different region of $((\Delta E)^2/\sqrt{D})\sqrt{t}$ and it looks mess. Thus we plot data in the way of Fig. 4(a). There the lines shows some distribution due to the distribution of M_0

and M_1, and also the effect of the dead time. However, we find that slopes of them are almost the same to each other and we estimate the constants as

$$\alpha_0 \simeq 3.5 \pm 0.1, \quad \text{and} \quad \beta_0 \simeq 2.3 \pm 0.1. \tag{20}$$

When we change ΔE, the value of M_0 and M_1 and the dead time change, but we find that the values of α and β are consistent although the scattering of date is larger.

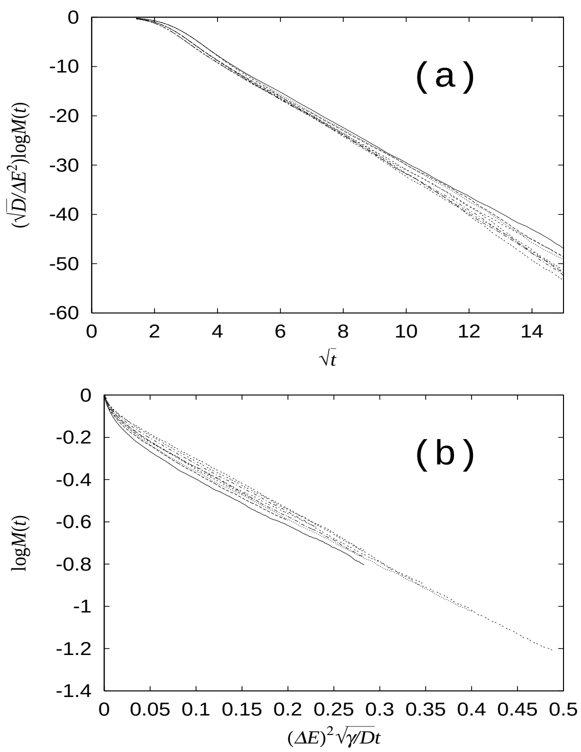

FIG. 4. Scaling plots (a)$(\sqrt{t}, \log M(t)/(\Delta E^2/\sqrt{D}))$, and (b)$(((\Delta E)^2\sqrt{\gamma/D})t, \log M(t))$

Finally we consider the case where the property of the noise depends on $M(t)$. Generally the change of the average magnetization causes the change of distribution of the field $h_{\rm ext}$ as discussed by Prokof'ev and Stamp [14]. In our analysis this change should be taken into account as a slow change of h_0, which may lead the same mechanism discussed in [14], which will be discussed in the future. Effect of change of the mean field to the LZS transition has been also discussed as a feedback effect which causes a large change of the transition probability [30]. This effect also causes important modification when the field is swept where the effective sweeping rate is modified. In the present case of fixed field, we assume that the time scale of the fluctuation field is much smaller than the change of the average magnetization.

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